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Application of lead monitoring results to predict 0e7 year old children's exposure at the tap

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abstract

Dwellings with/without a lead service line [LSL] were sampled for lead in tap water in Montreal, during different seasons. Short-term simulations using these results and the batchrun mode of the Integrated Exposure Uptake Biokinetic (IEUBK) model showed that children's exposure to lead at the tap in the presence of an LSL varies seasonally, and according to the type of dwelling. From July to March, for single-family homes, the estimated geometric mean [GM] blood lead level [BLL] decreased from 2.3e3.6 mg/dL to 1.5e2.5 mg/dL, depending on the children's age. The wide seasonal variations in lead exposure result in a minimal fraction (0e6%) of children with a predicted BLL >5 mg/dL in winter, as opposed to a significant proportion (5e25%) in summer. These estimations are in close agreement with the BLLs measured in Montreal children in fall and winter, and simulations using summer water lead levels illustrate the importance of measuring BLLs during the summer. Finally, simulations for wartime residences with long LSLs confirm the need to prioritize the control of this lead exposure from tap water.

1. Introduction

Neurodevelopmental effects have been measured in young children at low blood lead levels [BLLs], which makes it difficult to establish a safe threshold (Canfield et al., 2003; European Commission, 2011). Consistent with these findings, the Centers for Disease Control recently established a new reference value of 5 mg/dL based on population BLLs, and plans to review this value every four years (CDC, 2012). Children's exposure to lead [Pb] comes mostly from their diet but also from dust, soil, air, paint, and tap water. The reduction of Pb levels in food and gasoline resulted in the steady decline in BLLs in recent years. Also, remedial action has been performed to decrease the Pb levels in soil and dust at contaminated sites, although some of these sites still remain (USEPA, 2006). Considering the general decrease in Pb in these sources and the reduction of the BLL reference value, Pb in tap water may constitute the remaining significant contributor to children's exposure.

Lead service lines [LSLs] constitute a major source of Pb in tap water. Their replacement is expensive and legally complex

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Table 1 e Characteristics of tap water samplings performed in Montreal dwellings with/without an LSL.

Characteristics	Montreal campaigns (Deshommes et al., 2010; Cartier et al., 2011)			Health study campaign (Levallois et al., 2013)[a]
	2006	2007	2008	2009e2010
Dwellings (N)	109	45	59	306[b]
Dwelling type (N)	Wartime single homes with LSL (44); pre-1970 single homes with LSL (65)	Wartime single homes with LSL (11); pre-1970 single homes with LSL (34)	Single homes without LSL (27) or with LSL (wartime (7); pre-1970 (17); others (8))	Single homes (52); undetached dwellings in rows (44); duplexes, triplexes[c] (182); apartment buildings (28); with/without LSL
Samples (N)	451	152	284	2084[b]
Sampling type	30M stagnation; 5M flushing	RDT; 5M flushing	30M stagnation; RDT; 5M flushing	30M stagnation; 5M flushing; Bathroom random 250 mL sample
Sample type (N)	30M1L (109); 30M2L (109); 5M1L (109)	RDT1L (45); RDT2L (45); 5M1L (45)	30M1L (54); 30M2L (54); RDT1L (37); RDT2L (37); 5M1L (91)	30M1L (341); 30M2L (341); 30M3L (341); 30M4L (341); 30M5L (35); 30M6L (35); 30M7L (35); 30M8L (35); 5M1L (35); Bath250mL (223)[b]
Season	Spring (May, wartime homes); summer (JulyeSept, pre-1970 homes)	Summer (JulyeSept)	Spring (May, 7 homes without LSL); summer (JulyeSept)	Summer (Sept, N ¼ 16); fall (OcteNov, N ¼ 99); winter (DeceMar, N ¼ 226)
Temperature after 5 min of flushing (°C)[d]	Spring: 11.5 (10.1e12.7); summer: 21.6 (18.2e24.2)	21.6 (18.3e22.8)	Spring: 11.5 (10.7e12.7); summer: 22.3 (20.4e23.8)	Fall: 11.6 (7.9e21.5); winter: 3.6 (1.4e10.3); summer: 19.8 (17.4e21.7)
Sampling flow rate (L/min)[d]	5.5 (3.3e8.8)	6.0 (1.0e12)	6.7 (2.7e15)	5.6 (1.6e9.3)
PP volume (L)[d]	1.45 (0.13e6.46)	e	e	1.97 (0.12e5.83)[e]
LSL volume (L)[d]	1.98 (0.33e5.98)	e	e	e
LSL length (m)[d]	14.2 (2.60e30.2)	14.5 (6.00e30.2)	14.8 (6.0e27.5)	e

N e number.
a Measurement of Montreal children's BLLs.
b 306 dwellings sampled with 30M1L-4L and 5M1L sampling protocols (Sept2009eMar2010), 35 dwellings re-sampled with 30M1L-8L and 5M1L sampling protocols (NoveDec2010), either for LSL presence/absence confirmation (N ¼ 21) or re-confirmation (N ¼ 14).
c Two-family, three-family homes.
d Median (minemax).
e Determined on a subsample of 35 dwellings.

because of shared ownership between the municipality and the home owner. Flushing is a remediation method usually prescribed to reduce Pb levels below the 10e15 mg/L reference levels, although not a long-term solution. Also, even if LSLs are replaced, the benefits can be uncertain if the replacement is partial (Triantafyllidou and Edwards, 2011), or if long-term deposits were generated in the premise piping [PP] when the LSL was in place (Schock, 2005). Other sources include solders, brass fixtures, and faucets (Schock, 1990). Such sources, although containing far less Pb content than LSLs, can generate high Pb concentrations at the tap as well as Pb particles accumulations behind the tap, which could be highly relevant for exposure, especially in the case of large buildings (Deshommes and Prévost, 2012).

In several studies, an association has been observed between the presence of an LSL and increased BLLs in young children (Levallois et al., 2013; Brown et al., 2011; Edwards et al., 2009). The dissolution of Pb from an LSL is governed by water quality, and increases with stagnation time and temperature. Water temperature can vary widely across a distribution system and in PP, and may strongly influence Pb concentration at some taps (Schock, 1990). Regulatory Pb sampling is therefore often prescribed during summer, and consequently most of the Pb data available corresponds to this warmer period (Cartier et al., 2011; Deshommes et al., 2010). However, considering the seasonal variations of BLLs observed in children exposed to soil and dust mostly during the summer (Laidlaw et al., 2005; USEPA, 1996; Yiin et al., 2000), we suspect that similar trends in children's BLLs could occur following exposure to Pb in tap water. However, the extent of these variations has not yet been assessed.

The objectives of this study were: (i) to evaluate if results from tap water sampling can reveal the presence/absence of an LSL; (ii) to compare Pb sampling results for different seasons, and different types of dwellings with/without an LSL; (iii) to evaluate new sampling approaches and discuss their relevance for children's exposure; and (iv) to assess, using the Integrated Exposure Uptake Biokinetic [IEUBK] model, the effect of seasonal variations of Pb in tap water on the BLLs of young children living in homes with/without an LSL.

2. Materials and methods

2.1. Tap water samplings

Sampling for Pb in tap water was carried out in Montreal homes between 2006 and 2010. Table 1 summarizes the

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sampling approaches, which are further detailed in Deshommes et al. (2010), Cartier et al., 2011, and Levallois et al., 2013. In this paper, a single home is defined as a detached single-family home, with a service line that is not shared with neighboring dwellings. Also, summer corresponds to the JulyeSeptember period, fall to the OctobereNovember period, winter to the DecembereMarch period, and spring to the AprileJune period.

The 2006e2008 results originate from annual campaigns conducted by the City of Montreal, mostly during the summer, and generally in single homes with an LSL (Cartier et al., 2011; Deshommes et al., 2010). Two main types of single homes were sampled: (1) "wartime", which are small homes with a long LSL (median 20 m) constructed between 1940 and 1950; and (2) "pre-1970", which are homes constructed prior to 1970 with an LSL of average length (median 10 m), and that are not wartime. Several types of sampling were carried out at the kitchen tap, including: (i) 5M sampling by collecting a 1 L sample after 5 min of flushing; (ii) 30M sampling by collecting two 1 L samples after 30 min of stagnation; and (iii) random daytime [RDT] sampling by collecting two 1 L samples upon arrival at the home. During these sampling events, the presence/absence of an LSL was confirmed by visual inspection. Also, the volume and length of the LSLs and PP were determined for a significant number of the single homes visited (Table 1). Finally, a subset of four single homes with an LSL, including wartime homes, was followed over one year by profiling sampling consisting in collecting a 5M1L sample followed by six 1 L samples after 30 min of stagnation (30M1L-6L).

Conversely, 2009e2010 sampling was performed mostly during the fall and winter, in dwellings of multiple types, but in a relatively small proportion of single homes (Table 1). These samples were collected during a health study aimed at measuring the BLLs of 306 children in Montreal, as well as the Pb levels in their home environment, including in tap water, dust, and paint (Levallois et al., 2013). For the evaluation of tap water, 5M and 30M samples were taken at the kitchen tap on the same day as the BLL measurement. For the 30M samples, the first four liters were collected successively (30M1L-4L), with the objective of better assessing the Pb levels at the tap. Moreover, for a subsample of dwellings, a first flush sample of 250 mL (Bath250mL) was randomly collected at the bathroom tap. For this study, dwellings of multiple types located in older districts of Montreal were randomly selected, to increase the probability of including those with an LSL. However, the visual inspection of the service line and PP could not be performed. Therefore, the presence of an LSL was assessed by analyzing the Pb concentration profiles obtained at the tap (5M1L, 30M1L-4L). For households for which no conclusion could be drawn from these profiles, a second extended profiling sampling was performed later with eight liters rather than four (5M1L, 30M1L-8L), and with detailed inspection of the service line and PP. This extended sampling was also applied to a subset of 14 households already categorized with/without an LSL to double-check the validity of the LSL presence/absence determination method.

Finally, for all the dwellings sampled in 2007, and 2009e2010, the particles trapped in the tap aerator were collected and digested to estimate their total Pb content (Deshommes and Prévost, 2012).

2.2. IEUBK simulations

IEUBK is a validated model for representing exposure from various environmental sources containing Pb in 0e84 month old children. Parameters relative to each of the sources and to children's exposure (ingestion rates, Pb concentrations, etc.) are entered into the model, which estimates a geometric mean (GM) BLL, and a proportion of children having a BLL over a threshold selected by the user (5mg/dL in this study). Version IEUBKwin1.1_Build11 was used, and exposure to Pb in tap water, soil, dust, air, and diet were considered. The model entry parameters are detailed in the Supporting Information [SI] section. No differentiation was made between the dissolved and the particulate fraction of Pb in tap water, since only the total Pb was measured during the 2009e2010 samplings, and most of the particulate Pb results from traditional sampling in Montreal homes were low (Deshommes et al., 2010). Therefore, total Pb in tap water was considered as soluble Pb and 50% bioavailable, in agreement with the high absorption rate of children for soluble Pb forms (Mushak, 1998). For all the simulations, median total Pb concentrations from the 2006e2010 samplings were applied. Short-term simulations for representing seasonal exposure to Pb levels in tap water were performed using the IEUBK batchrun mode, according to Donohue et al. (2011). This mode, using an Excel shell linked to IEUBK via Visual Basic, makes it possible to vary the tap water exposure concentration for each month of the children's age, from 0 to 84 months.

3. Results and discussion

3.1. Evaluation of the presence of an LSL

Typically, the presence of an LSL can be inferred from the progressive increase in Pb concentrations in several successive liters collected at the tap following stagnation (Giani et al., 2005). It is then concluded that the liters with markedly higher concentrations correspond to the liters from the service line. Therefore, depending on the PP and LSL volumes, the liters with increased concentrations among the samples collected

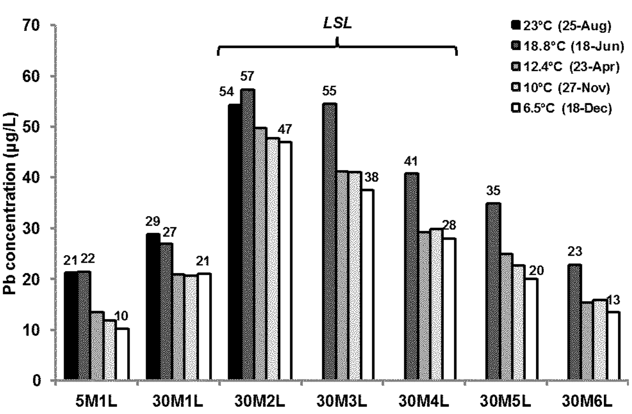

Fig. 1 e Typical profile of Pb concentrations at the kitchen tap of a wartime single home with an LSL in Montreal, for different tap water temperatures (measured after 5 min of flushing).

will vary. Fig. 1 presents the profile in one of the wartime single home sampled at various water temperatures (6.5e23 °C). Even after 5 min of flushing (5M1L), Pb levels exceed 10mg/L, which clearly indicates the presence of an LSL. After a short stagnation period, first flush 30M1L sample concentrations ranged from 21 to 29 mg Pb/L. Lead levels markedly increased in the two subsequent samples (30M2L-3L), and then progressively decreased in the 30M4L-6L samples to the background levels measured in the 5M1L samples. These peak concentrations in the profile indicate that the 30M2L-3L samples correspond to the volume of water from the LSL. Also, although Pb concentrations decrease with decreasing tap water temperatures in December (6.5 °C), noticeably higher concentrations are still measured in the 2nd and 3rd liters (Fig. 1). These clear trends show that concentration profiling can easily be interpreted to confirm the presence of an LSL in single homes, even in cold water temperatures. The impact of temperature on Pb release was verified over a year in four single homes. Results showed that lower temperatures depressed the median total Pb concentrations 2.9-fold for 5M1L samples, and 2.2-fold for 30M1L-2L samples [SI].

Interpretation of the Pb concentrations is essential for the households when no visual inspection of the service line is performed, which was the case during the health study. Lead concentration profiles were more challenging to interpret for these households as compared to the four single-homes mentioned previously, considering the generally low Pb concentrations measured (90th percentile of 7.4mg/L). Most of the sites sampled were multiple dwelling homes. Therefore, more than one dwelling can be connected to a single LSL, and the water use restrictions required to ensure controlled stagnation during sampling are more difficult to implement and control. In addition, the PP volume from the kitchen tap to the service line for some dwellings can exceed four liters (Table 1). Therefore, in some cases, the first four liters may not be indicative of concentrations in the service line, and therefore do not allow for the detection of an LSL. The interpretation of the 30M1L-4L profiles was even more challenging because of the cold water temperature (median of 3.6 °C in 5M1L samples). Based on a thorough examination of the Pb concentration profiles, a decision tree was built to sort the dwellings according to the probability of the presence of an LSL in cold water (Fig. 2). As the 1st liter collected at the tap mainly reflects the PP contribution, only 30M2L-4L and 5M1L samples were considered for the sorting. First, it was established that a Pb concentration ≥ 1mg/L in a flowing 5M1L sample indicated a high probability of the presence of an LSL. In this case, if the concentration in at least one of the 30M2L-4L samples exceeded 1.5mg/L, we concluded that an LSL was present. Else, an LSL was probably present (results inconclusive). If, by contrast, the Pb concentration in 5M1L samples was <1 mg/L, then the probability of the presence of an LSL was considered low. In those cases, if the concentration in at least one of the 30M2L-4L samples exceeded 1.5mg/L, then it was probable that an LSL was present (results inconclusive), otherwise it was considered absent. Therefore, 30M2L-4L samples ≥ 1.5 mg/L were considered indicative of the presence of an LSL (Fig. 2). Applying this decision tree resulted in the classification of all but 30 of the 306 dwellings in the health study (probable LSL). The second extended profiling sampling was performed for 21 of these 30 homes (8 could not be re-sampled, and for the last one, the owner confirmed the presence of an LSL by phone) in NovembereDecember 2010 (7.1e12.3 °C). The volume of the PP and service line were thoroughly measured during this extended sampling (Table 1), and used to interpret the concentration profiles. If the concentration increases coincided with the volumes from the service line, it was concluded that an LSL was present, otherwise it was deemed that no LSL was present [SI]. Using the results from the decision tree and from the extended profiling sampling, it was concluded that in the 306 dwellings sampled in the health study, 171 were served by

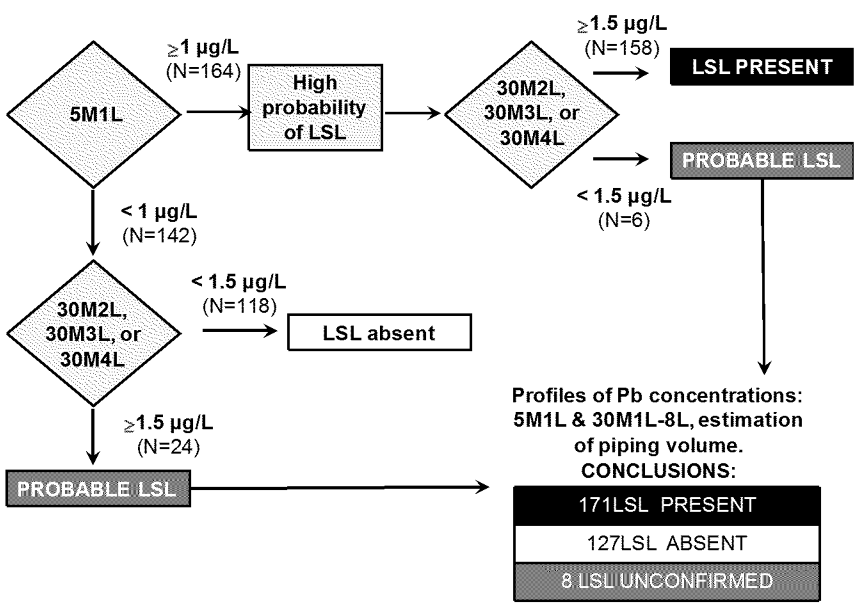

Fig. 2 e Decision tree for determining the presence of an LSL according to the Pb concentrations measured at the kitchen tap, in cold water.

an LSL, 127 were not served by an LSL, and 8 were unclassifiable (Fig. 2). Finally, 14 households already confirmed with or without an LSL by the decision tree were re-sampled using the extended profiling sampling. These households were selected to test multiple situations of the decision tree and its boundaries, and the detailed data confirmed the prior classification with the decision tree. Overall, when considering all results from the health study, Pb concentrations were significantly lower in households categorized as being without an LSL as compared to those categorized as having an LSL (KruskaleWallis test, p < 0.01).

3.2. Effect of water temperature and dwelling type on Pb concentrations

Lead concentrations at the kitchen tap were segregated by sampling type, sampling campaign (Montreal campaigns/ health study) and dwelling type, according to the presence (Fig. 3) or absence (Fig. 4) of an LSL. Samples collected mostly during the winter, in dwellings with an LSL showed lower Pb concentrations (Fig. 3b; 0.3e61 mg/L; health study), as compared to those collected mostly during the summer (Fig. 3a; 2.1e249 mg/L; Montreal campaigns). This effect is partly explained by the impact of temperature on Pb dissolution from LSLs in this system (Cartier et al., 2011). As well, the Pb levels in the 5M samples collected from homes with an LSL during the health study were correlated to the water temperature [SI]. The type of dwelling also impacted Pb levels measured during the health study. As mentioned previously, only 52 single homes were sampled, and 1.2e21 mg Pb/L (10the90th percentile for all samples) was measured in these homes compared to 1.4e7.9 mg Pb/L for multiple dwellings (Fig. 3b). Moreover, the Pb concentrations measured in the different types of homes were significantly different from one another, whether the fall and winter results were considered separately or together (KruskaleWallis test, p < 0.01). The single homes with LSLs from Montreal campaigns also show clear differences, as the Pb levels in the 30M1L-2L samples were significantly higher for wartime homes sampled in the spring (12 ¹C, median 22 mg/L) than for pre-1970 homes sampled in the summer (22 ¹C, median 13 mg/L) (Fig. 3a; KruskaleWallis test, p < 0.01). This was explained by the typically long length of the LSL supplying the wartime homes (Cartier et al., 2011). Finally, a 90th percentile of 43 mg/L was measured for pre-1970 homes sampled with RDT as compared to 32 mg/L with 30M samplings (Fig. 3a), and reflect the differences in sampling protocol (Deshommes et al., 2010).

Pb concentrations were considerably lower in dwellings without an LSL (Fig. 4) than in those with an LSL (Fig. 3). For the Montreal campaigns results, based on the median for each sample type and for each dwelling type, the Pb concentrations were 8.5e29 mg/L lower in dwellings without an LSL as compared to dwellings with an LSL (Figs. 3a and 4a). Smaller differences (1.7e11 mg/L) were measured during the health study (Figs. 3b and 4b), although they were more marked for single homes considered solely (3.7e11 mg/L). Therefore, in Montreal, the concentrations at the tap in dwellings with an LSL will vary according to water temperature and dwelling type. The children participating in the health study were therefore exposed to lower Pb concentrations in their tap water, relative to the Pb levels measured in single homes in the same distribution system in summer. The BLLs measured are nevertheless a valid measurement of the exposure of many children to Pb in tap water in Montreal, since colder water temperatures last for about 4e5 months a year, and multiple dwellings are common (>70% of all 1e8 family homes in Montreal considered in this situation based on the City's data).

Finally, the results obtained for dwellings without an LSL, for winter (health study) and summer (Montreal campaigns) (Fig. 4), were comparable, considering the 30M2L and 5M1L results (ᴶ 0.4 mg/L based on medians). This similarity of Pb concentrations strengthens the validity of the decision tree regarding households' classification from the health study. Moreover, no significant correlation was found between the 5M1L results and water temperature [SI]. In fact, for these dwellings, PP is the major source of Pb, and is exposed to warm water during both summer and winter, through home

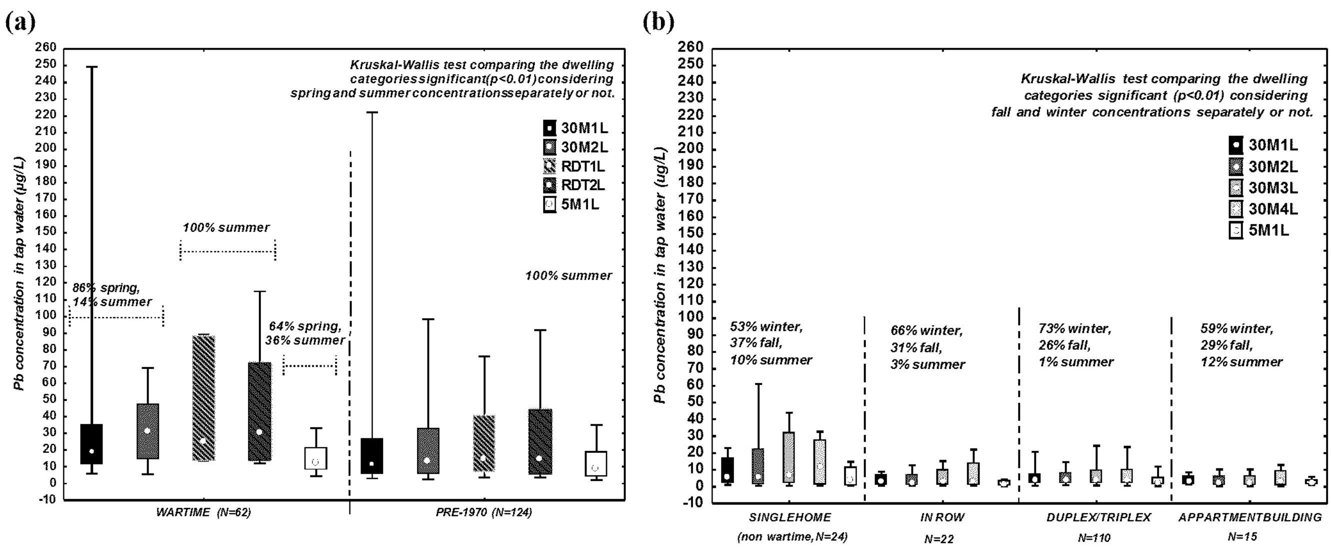

Fig. 3 e Pb concentrations at the kitchen tap in dwellings with an LSL per dwelling type: (a) Montreal campaigns (spring, summer); and (b) health study (fall, winter). Median (10the90th percentile) box-plot, minemax whiskers.

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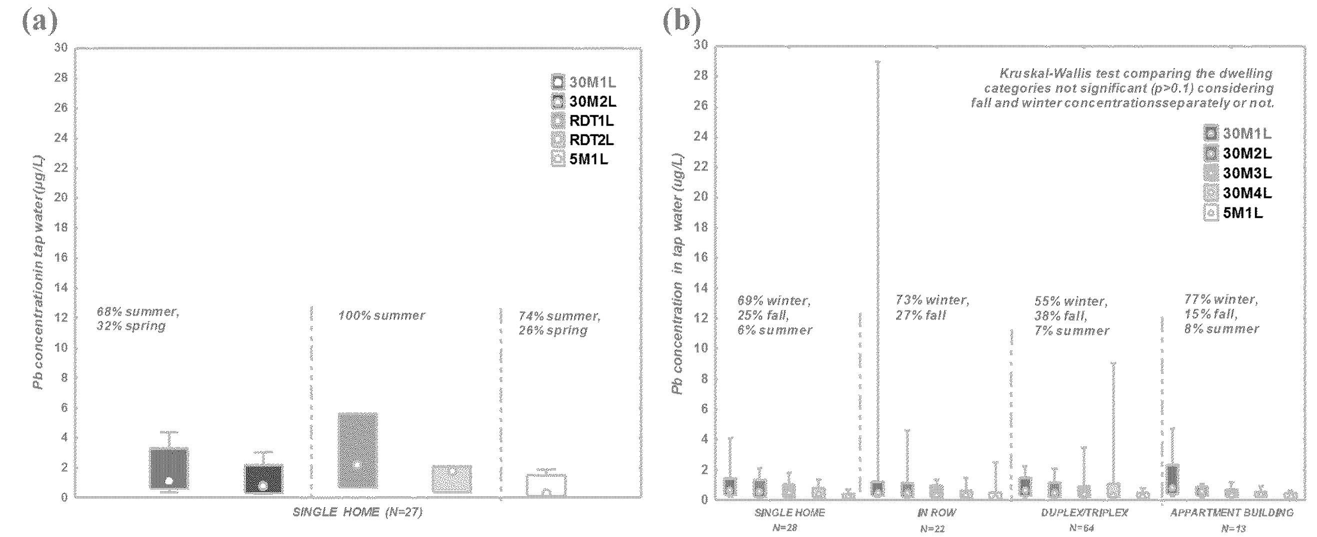

Fig. 4 e Pb concentrations at the kitchen tap in dwellings without an LSL per dwelling type: (a) Montreal campaigns (spring, summer); and (b) health study (fall, winter). Median (10the90th percentile) box-plot, minemax whiskers.

heating. Moreover, median Pb levels in the 30M1L samples are a bit higher in Fig. 4a (1.2 mg/L), compared to those in Fig. 4b (0.5e0.8 mg/L), reflecting differences in PP/faucet. Finally, no significant difference was found between the Pb levels and the dwelling types in the absence of an LSL (KruskaleWallis test, $p > 0.1$). Therefore, the effects of water temperature and dwelling type on Pb levels are clearly only observed in the presence of an LSL.

3.3. Comprehensive in-home sampling

Regulated sampling protocols usually prescribe sampling the kitchen tap at a low flow rate, with the aerator in place. However, children may also be exposed to Pb from tap water from other taps in the home. The first flush of 250 mL was

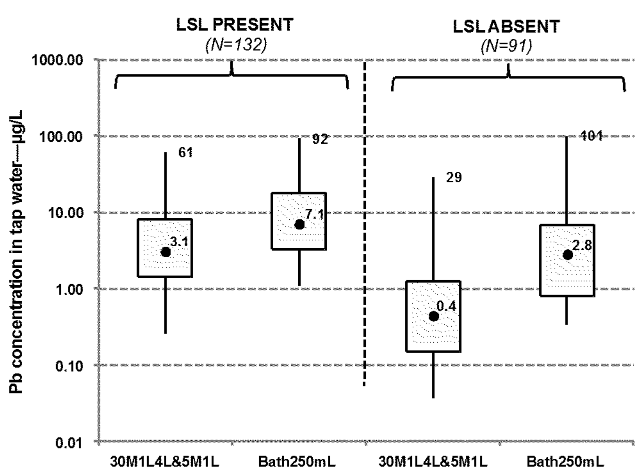

Fig. 5 e Pb concentrations at the bathroom tap (Bath250mL) and Pb concentrations for 30M&5M samples in the corresponding dwellings (health study). Median (10the90th percentile) box-plot, minemax whiskers.

collected randomly at the bathroom tap of 223 homes during the health study. Bathroom tap samples, in the presence or absence of an LSL, showed significant variations in total Pb levels: medians and 90th percentiles varied within 2.8e7.1 mg/L and 6.8e18 mg/L respectively (Fig. 5). Although the 90th percentile concentrations were slightly higher in the presence of an LSL, comparable maximum values were measured with/without an LSL (92, 101 mg/L). The background concentration in the presence of an LSL (30M1L-4L&5M1L in Fig. 5) is the contribution from the LSL during flow, with additional Pb resulting from dissolution occurring during stagnation. For dwellings with an LSL, these background levels appear to be higher than the contribution from the tap, although peak concentrations may be related to the PP. These results indicate that Pb concentrations at a bathroom tap can be higher than at the kitchen tap. This may be explained by the type of faucet and differences in PP configuration upstream of the faucet (Triantafyllidou and Edwards, 2012), but also by the sample volume used (250 mL). Seasonal water temperature was not determined to be a cause of these higher concentrations, since the bathroom samples were representative of the faucet sampled and its upstream PP, and no significant difference was found between samples collected in the fall and in the winter (t-test, p ¼ 0.26). Considering that these taps are usually easily accessed by children, and that children may not know which faucets are safe for drinking, bathroom tap water may possibly be a route of Pb exposure to consider for children. Additional studies are needed to evaluate the consumption patterns of children at such taps, so that this exposure can be adequately considered in exposure models.

Sampling flow rate is another factor to consider in exposure assessment. The results presented in Fig. 3 were collected at low to normal flow rates (5.7 ┘ 1.4 L/min). These conditions are not typical of consumer consumption patterns, and can decrease Pb concentrations at the tap (Deshommes et al., 2010). It may be important to consider results from high flow rate sampling, to avoid underestimating children's exposure and to refine exposure estimates. Observations suggest that recommendations on sampling flow rate, and the use of wide-mouth sampling bottles are needed, so that the samples can be adequately collected (Triantafyllidou et al., 2007).

Finally, another aspect of exposure studied was the characterization of the particles trapped in the tap aerator. Results indicated 0e71% Pb (weight %) in all the sets of particles collected (median 4.7%), whether or not an LSL was present, since these particles originated mostly from PP (Deshommes and Prévost, 2012). Such particles can increase Pb levels at the tap by dissolving or by breaking into smaller particles (Cartier et al., 2011). The extent of exposure due to these particles is however difficult to characterize. Regardless, limiting the exposure to these particles by flushing the tap or by cleaning the faucet aerator regularly would be beneficial. Furthermore, such preventive measures can be easily communicated and implemented.

3.4. IEUBK simulations e impact of sampling protocol, season, and dwelling type

IEUBK simulations were first run in normal mode, using a fixed Pb concentration of exposure in tap water. Median concentrations were used as input for exposure to tap water, and were segregated by sampling type, season, presence/absence of an LSL, and dwelling type (Fig. 6). As no significant difference was observed among dwellings without an LSL, the results from all dwellings were combined in the simulations in this case. Each of the Pb concentrations applied combined results from 2006 to 2010, although some sampling types were particularly performed in a specific year (Table 1). Also, when the 30M results from 2006 to 2010 were grouped together, the medians from the 30M1L-2L and 30M1L-4L samples were calculated, and the highest value was selected for the simulations. It should be noted that no samples were taken after prolonged stagnation (>30 min).

Results from dwellings without an LSL, represented in grey in Fig. 6a, are quite similar to the estimation obtained without any contribution of tap water (background contributions from soil, dust, air, and diet). Regardless of the sampling protocol used or the season, the estimated GM BLL varies in the 1.2e1.4 mg/dL range, and only 0.1e0.3% of children are estimated to present a BLL >5mg/dL. Considering the results from single homes with an LSL (excluding wartime homes), represented in black (Fig. 6a), the first general observation is the seasonal decrease in BLLs in cold water, which is supported by the lower Pb results presented in Fig. 3. Using the 30M sample results, the estimated GM BLL falls from 3.1 in summer to 2.1mg/dL in winter. As well, the estimated fraction of children with a BLL >5mg/dL decreases from 15% to 3.0% based on the 30M results, and from 7.3% to 1.1% using the 5M results (Fig. 6a). Our second observation is the impact of the sampling protocol on the estimation of exposure. Indeed, the estimated exposure for summer, fall, and winter in single homes with an LSL decreases depending on the sampling protocol applied, in the following order: 5M < 30M < RDT. Clearly, for these specific homes, flushing for 5 min decreases the estimated BLLs, however flushed samples are not representative of a realistic consumer tap exposure. When using results from sampling protocols which yield results that are more indicative of typical concentrations at the tap (30M, RDT), the BLLs calculated reach 3.1e3.4 mg/dL, and 15e19% exceed 5 mg/dL in summer. In conclusion, Pb concentrations at the tap and estimated exposure vary considerably with water temperature and sampling protocol. These findings show that the selection of a protocol that is the most representative of typical concentrations at the tap is important for the evaluation of tap water contribution to children's exposure in warm water. Van den Hoven and Slaats (2006) have shown that RDT, and to a lesser degree 30M sampling, provides the closest estimation of weighted proportional sampling concentrations measured at consumers' tap. Our simulations also reveal the significant underestimation associated with the use of flushed samples to estimate the impact of tap water on exposure.

Results segregated by type of dwelling with an LSL, by season (for which results were available), and by sampling protocol (30M, 5M) show a different trend depending on the dwelling type (Fig. 6b). First, when comparing similar seasons (fall and winter), predicted BLLs are slightly higher for single homes (1e6%; 1.7e2.4mg/dL) than for other types of dwellings (0e4%; 1.3e2.2 mg/dL), reflecting the higher tap water concentrations in these homes (Fig. 3b). Also, the impact of the sampling protocol on exposure observed for single homes in Fig. 6a is barely noticeable for in row dwellings and apartment buildings sampled in fall and winter. A small decreasing trend is observed in fall for two-family/three-family dwellings (duplex/triplex) between 30M and 5M samplings; however, this effect is not seen in winter. This may be explained partly by the cold water temperatures, but also by the dwelling configuration (shared LSL) and the consumption patterns in these specific dwellings (number of people using water from the same LSL). In these dwellings, the estimated GM BLL (1.3e2.2 mg/dL) and the estimated fraction of children with a BLL >5mg/dL (0e4%) do not vary excessively with the sampling protocol (30M, 5M) or the season (fall, winter). Clearly, the sampling protocol has little impact on the estimation of the exposure through tap water during cold periods for these dwellings; however, the impact is greater for single homes. Nonetheless, summer sampling data would be valuable for confirming the low levels of exposure in these dwellings.

For wartime homes in spring (green color on Fig. 6b) and summer, simulated BLLs show that the children living in such dwellings are considerably more exposed than those living in other types of dwelling. Based on the 30M sampling data, the estimated GM BLL varies within 4.4e4.6 mg/dL, and about 38e42% of children are estimated to have a BLL >5mg/dL. Most importantly, even when using the most lenient estimate of tap water exposure by applying the median concentration after 5 min of flushing, the GM BLL is estimated at 2.9e3.8 mg/dL, and the proportion of children with BLL >5 mg/dL reaches 12e27%. Flushing prior to consumption is commonly recommended by public health authorities and utilities to consumers living in a house with an LSL. However, even though extended flushing does reduce the median concentrations at the tap, this action is clearly insufficient as 27% of the children exposed to these concentrations may still exceed the 5 mg/dL threshold. Our results show that specific types of single homes, such as wartime homes served by long LSLs, represent a worst case scenario when compared to other types of dwellings, even other single homes with an LSL.

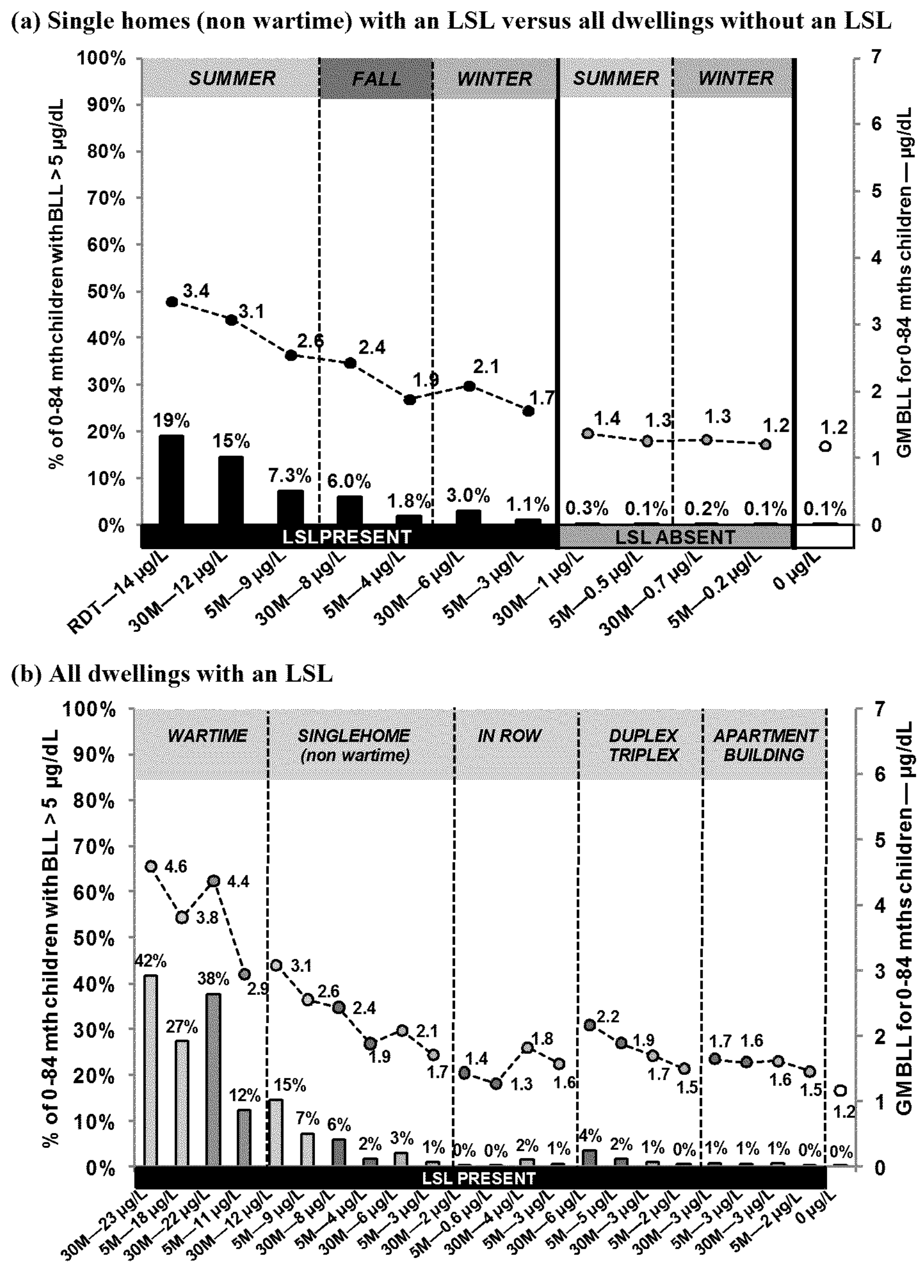

Fig. 6 e IEUBK estimates for a fixed Pb concentration of exposure in tap water (median value) per sampling type and per season: (a) for single homes non wartime with an LSL and for all dwellings without an LSL; and (b) for all dwellings with an LSL.

3.5. IEUBK simulations e impact of seasons

Based on the seasonal variations of Pb in tap water (Fig. 3), and their impact on the IEUBK simulations for each season (Fig. 6), the batchrun mode in IEUBK was used to study the effect of short-term increases of Pb concentrations at the tap. Considering the limited data for summer for dwellings with an LSL other than single homes, simulations were run only for single homes with an LSL (wartime/non wartime). Also, as the dwelling type showed no impact on Pb levels in the absence of an LSL, all the dwellings were combined for simulations performed without an LSL. The simulations considered that the child was born in January (Jan-1), and that the child's seasonal pattern of yearly exposure was repeated up to the age of 7 (Dec-7). A 50/50 weighted average of median concentrations from 30M&RDT (combined) and 5M samples was calculated for each season, in order to represent a realistic scenario of moderate exposure. The weighted average for the winter was then applied from December to March of each year, while the weighted averages for spring, summer, and fall were applied for the AprileJune, JulyeSeptember, and OctobereNovember periods respectively. Since no spring data were available for single non wartime homes, the fall values were used. Also, only spring and summer data were available for the wartime homes. For these specific dwellings, fall and winter data were estimated from the subset of four homes followed over the

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year with profiling sampling, which showed comparable Pb levels for spring and summer [SI].

Fig. 7 shows the seasonal variations in BLLs and the corresponding proportion of children with BLLs >5 mg/dL. For homes without an LSL, the IEUBK results closely follow those obtained without any contribution of tap water, revealing that tap water does not increase BLLs significantly. The estimated GM BLL varies in the 0.77e1.8 mg/dL range and remains quite stable year-round, and, as expected, globally decreases with the children's age. Conversely, the simulations for single

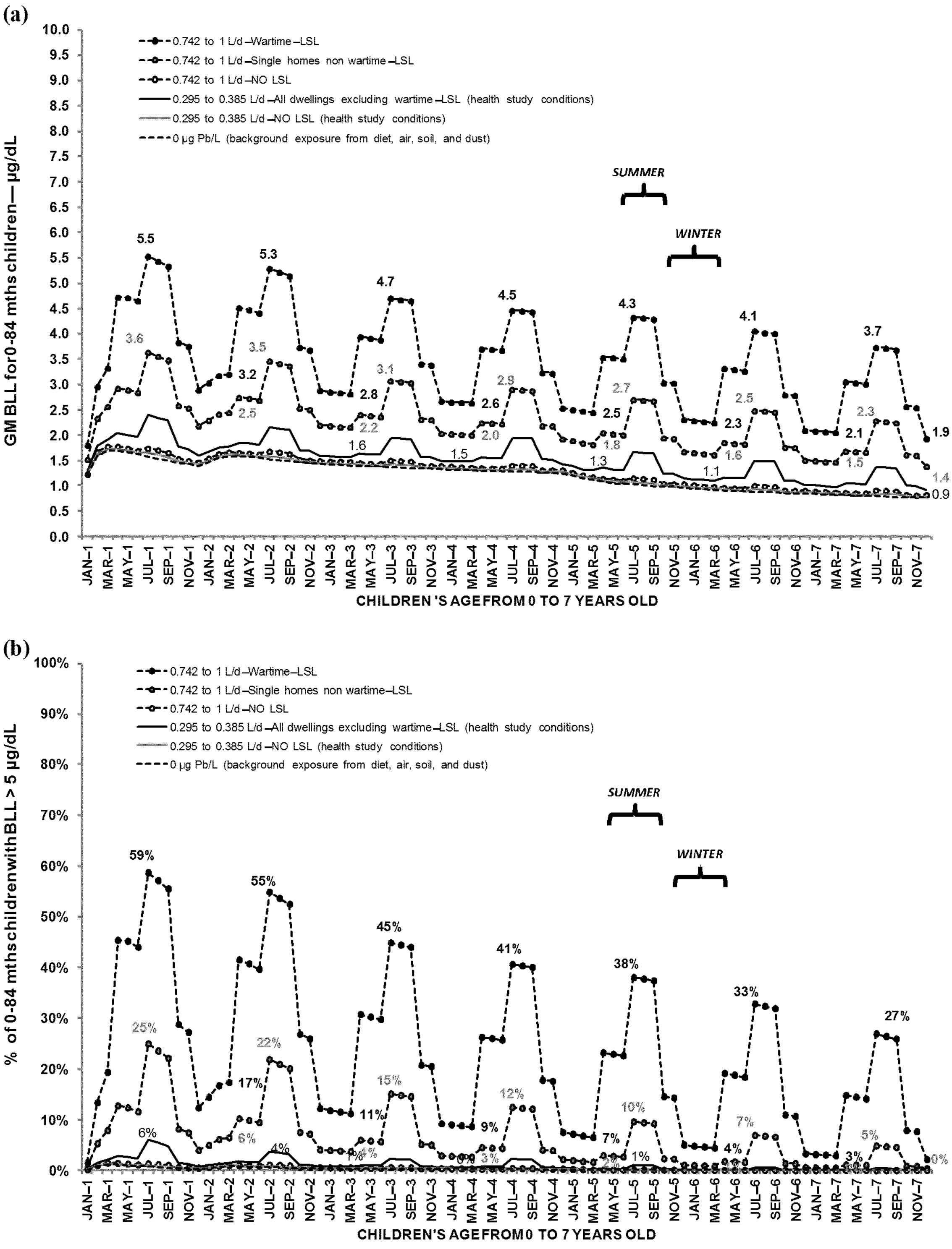

Fig. 7 e IEUBK estimates of: (a) the GM BLL; and (b) the proportion of children with a BLL > 5 mg/dL, from 0 to 84 months age (0 months corresponding to JAN-1). Figures for single homes with an LSL (wartime or non wartime) and for all dwellings types without an LSL, applying seasonal Pb concentrations for tap water. Seasonal data are a weighted means of 50% of the median from the 30M&RDT samples, plus 50% of the median from the 5M samples collected in the dwellings for the season concerned.

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homes with an LSL predict higher BLLs overall, with marked seasonal variations. The estimated GM BLLs increase from about 1.5–2.5mg/dL in March, depending on the children's age, to about 2.3e3.6mg/dL in July. For wartime homes, these levels vary from 2.1–3.2mg/dL to 3.7e5.5mg/dL (Fig. 7a). Children from the health study ranged from 1 to 6 years old, but 62% were 3e6 years old (Levallois et al., 2013). During the health study, performed mostly during the winter, a GM BLL of 1.2mg/dL was measured for children living in homes without an LSL, while a GM BLL of 1.5mg/dL was measured for children living in homes with an LSL, with all ages combined. Our IEUBK estimations for homes without an LSL (0.77e1.8 mg/dL) fall within the 1.2mg/dL GM measured in Montreal children. Conversely, the BLLs modeled for single homes with an LSL in winter (1.5e2.5 mg/dL) exceed the measured BLLs. The observed difference may be explained by the entry parameters applied in IEUBK, particularly the relatively high daily water intake (742e1000 mL/d). This choice is justified considering the consumption values used to estimate children's exposure to Pb from tap water by the local public health authorities and recent European guidelines (Beausoleil and Brodeur, 2007; European Commission, 2011). In addition, comparable values were recently measured for Canadian children <5 years old (Jones et al., 2006). Children from the health study belonging to a subpopulation of educated families were shown to drink small volumes of tap water (295e385 mL/d, questionnaire). Secondly, BLLs from single homes reflect higher Pb concentrations at the tap than those measured in the other types of dwellings in which the children from the health study were living. For comparison purposes, simulations were run using Pb levels from all dwellings in the health study (winter, fall, and spring values), Pb levels from single non wartime homes from the Montreal campaigns (summer), and the low consumption volumes from the health study. Using these entry values, the estimated GM BLL for 1e6 year old children in winter varies in the 1.1e1.6mg/dL range, which falls within the measured BLLs (1.5mg/dL, Fig. 7a). The slight difference could also be caused by: (i) the background levels applied for soil, dust, diet, and air that were not specific to the study area, and (ii) the age range of children from the health study (62% were >3 years old). Furthermore, in the health study, the BLLs for approximately 25% of the 1e6 year old children exceeded 1.8mg/dL, and for 5% of the children, the BLLs exceeded 3.1mg/dL. It is interesting to note that, even when lower consumption volumes from the health study are used, a small proportion of children with BLLs >5mg/dL remains even in winter (0e1%, Fig. 7b). Considering the differences in baseline exposure to sources other than tap water, these trends are considered to be consistent.

The seasonal variations in BLLs in dwellings with an LSL are even more noticeable when the estimated proportion of children with a BLL >5 mg/dL is considered. Indeed, when considering single homes, but excluding wartime homes, the range increases from 0–6% in March to 5e25% in July, depending on the children's age (Fig. 7b). For wartime homes, these ranges are systematically higher: 3e17% in winter and 27e59% in summer. Finally, when applying Pb levels in all dwelling types combined and the lower daily intakes from the health study, this range is estimated to drop to about 0e1% in winter and 0e6% in summer. This distribution of BLLs is set by the geometric standard deviation (GSD) value of 1.6 applied by default in IEUBK, which takes into account the various sources of variability in children's exposure (biological and behavioral differences, spatio-temporal variability of Pb concentrations, measurement and analytical variability). This value drives the BLL distribution and sets the estimation of the proportion of children in whom a given threshold value is exceeded. It is possible that this value does not adequately reflect the study site conditions. However, the IEUBK guidance manual specifies that the GSD of 1.6 was derived from actual BLL distributions from several neighborhoods, and that it applies to most of sites. Using an alternate GSD value should only be considered with supporting evidence. Based on our estimations using the recommended GSD of 1.6, particularly when considering dwellings other than wartime homes, the distribution shows the low probability of detecting children with a BLL >5mg/dL during the winter in colder climates, such as in Montreal, unless a large sample size is used. These estimates also show that BLL studies should be conducted in summer to detect seasonal exceedances. Similar seasonal variations have been reported by epidemiological studies on soil and dust exposure, since exposure to Pb from these sources is enhanced in summer, due to higher ingestion rates. Indeed, children BLLs increased by þ1.0e4.0 mg/dL in summer, compared to winter in Indianapolis, Syracuse, and Jersey City, due to increased exposure to soil/dust (Laidlaw et al., 2005; Yiin et al., 2000). In addition, the USEPA (1996) reported BLLs about 40% higher in summer than in winter, due to increased exposure to outdoor Pb sources, the increases being the highest for 2e3 year olds (active hand-to-mouth activity). These variations are not modeled by IEUBK unless an Excel shell is developed to vary monthly children's ingestion rates of soil/dust. The public health consequences of short-term exceedance of the 5 mg/dL BLL should be evaluated, to determine the importance of implementing remedial action for seasonal sources.

For our simulations, background levels of soil/dust concentrations were used, and only tap water concentrations varied seasonally. Although the ingestion rates applied for the 2e4 years old were the highest of all the age categories, the BLLs estimated for summer in dwellings with an LSL for this age group could potentially be higher, considering the probable simultaneous increase in exposure to Pb in tap water, soil, and dust. Finally, the IEUBK estimations for wartime homes were always the highest, the proportion of children with a BLL >5 mg/dL being about twice that estimated for children living in single homes with an LSL (Fig. 7b). Specifically, for these homes in Montreal, it would be a matter of priority at short-term to reinforce the public health recommendations concerning alternative sources of drinking water (bottled water, point-of-use devices) to limit the exposure of young children and pregnant women at the individual level, and for the municipality to study corrective measures that could be implemented (corrosion control, LSL replacement).

Conclusion

The detection of an LSL based on a Pb concentration profile is possible, although easier to perform in single homes during the summer. This requires a good understanding of the evolution of Pb concentrations with water temperature, and of the dwellings' type and configuration. In Montreal, Pb concentrations at the tap decrease considerably with a drop in water temperature, obscuring the differences that might be present due to sampling protocol. However, in warmer temperatures, the concentrations after stagnation are higher, resulting in differences in exposure assessment among the various protocols. Therefore, the selection of the sampling protocol is more important in warm water.

In dwellings supplied with an LSL, Pb in tap water contributes to children's exposure. This contribution is the highest in summer, in single homes, and is exacerbated in wartime homes in the case of Montreal. This contribution also depends on children's consumption pattern, which is reflected by the sampling protocol. For children drinking tap water and living in dwellings served by an LSL, tap water would be one of the dominant sources of exposure. For children also exposed to soil and dust via hand-to-mouth activity, the estimated exposure could possibly be higher during summer, considering that soil/dust intakes are generally greater during that period.

For dwellings with an LSL, the IEUBK estimations overestimated the GM BLL measured in Montreal children during the health study conducted in fall and winter if reference daily tap water consumption values were used, as is the case in regulatory risk analysis. Modeled BLLs for fall and winter closely matched measured BLLs (| 7%) when lower reported water consumption volumes were used, showing the importance of selecting important yet realistic daily consumption volumes when evaluating tap water contribution. The simulations also revealed that measuring the BLLs of children living in homes with an LSL during the cold season may miss a certain proportion of children exceeding 5 mg/dL. However, results are reassuring as they indicate that children are much less exposed to Pb in tap water during the cold season. Results also point out that Pb concentrations in dwellings other than single homes contribute less to exposure, and that LSL replacement in these homes may be a lower priority than in single homes. Nonetheless, summer data would be needed to confirm the Pb concentrations in such multiple dwellings. In the case of Montreal, wartime homes clearly represent the highest level of exposure to Pb at the tap. For these homes, existing advisories should be strengthened and the short-term total replacement of these LSLs should be a priority.

Considering the Pb concentrations at the bathroom tap, and the particle accumulations behind the tap aerator, specific recommendations should be issued to limit exposure, whether or not there is an LSL delivering the water. Finally, regulating high flow rate samplings and applying their results in IEUBK would be needed to reflect consumer usage and to refine the estimation of children's exposure.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.watres.2013.02.010.

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